

Glassy dynamics: effective temperatures and intermittencies from a two-state model

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We show the existence of intermittent dynamics in one of the simplest model of a glassy system: the two-state model, which has been used¹ to explain the origin of the violation of the fluctuation-dissipation theorem. The dynamics is analyzed through a Langevin equation for the evolution of the state of the system through its energy landscape. The results obtained concerning the violation factor and the non-Gaussian nature of the fluctuations are in good qualitative agreement with experiments measuring the effective temperature and the voltage fluctuations in gels and in polymer glasses. The method proposed can be useful to study the dynamics of other slow relaxation systems in which non-Gaussian fluctuations have been observed.

I. INTRODUCTION

Complex systems are often distinguished by the existence of a very intricate free energy landscape consisting of many barriers which the system has to overcome to evolve. It is precisely the presence of these barriers the responsible for the slow relaxation dynamics which manifests in the appearance of peculiar phenomena as aging, lack of a fluctuation-dissipation theorem and intermittencies caused by the presence of large fluctuations. These features, predicted and observed in systems of different nature as glasses, granular flows, foams, crumpled materials and in the dynamics of the disordered systems^{2,3,4,5,6,7}, have attracted the interest of many researchers during the last years with the purpose of describing the main features of slow relaxation dynamics⁸.

In a previous paper¹, we have proposed a minimal relaxation model aimed at characterizing the dynamics of a system relaxing in two very different time scales, which are related to inter-well and intra-well relaxation processes. Two scenarios were analyzed for this purpose. In the first of them, the system may explore the whole reaction coordinate space undergoing a diffusion process described by a Fokker-Planck equation⁹, which accounts for the intra-well and inter-well relaxations. In the other, obtained from the first one by eliminating the fast variable, the system undergoes an activated process. In spite of its simplicity the model shows some of the peculiar features of the dynamics of slow relaxation systems and proposes an explanation of why and how the fluctuation-dissipation is violated. It was found that the violation factor or effective temperature depends on the observable and on the initial populations in the wells. This result shows that the effective temperature does not univocally characterize the thermal state of a glassy system undergoing activated dynamics¹⁰.

Our purpose in this paper is to use that model to explain the presence of intermittencies in the dynamics of

a system in a glassy phase and the non-Gaussian nature of the probability distribution function, which have recently been observed in measurements of the dielectric properties of gels and polymer glasses^{11,12}, and in some theoretical studies of spin-glass models^{13,14}.

The paper is organized as follows. In Section 2, we analyze some of the main traits of glassy dynamics from a two-state model. Section 3 is devoted to present the results concerning the intermittent behavior and the non-Gaussian nature of the fluctuations. Some conclusions and perspectives are presented in the final section.

II. GLASSY DYNAMICS FROM A TWO-STATE MODEL

In a two-state model, the minimal relaxation model for glassy systems^{15,16,17}, one assumes that the process consists of two main steps: a slow relaxation, in which the coordinate characterizing the state of the system jumps from a potential well to the next one, and a fast equilibration process in the well. It has been shown¹ that the dynamics of the system can be analyzed in terms of a Fokker-Planck equation describing a diffusion process through the free energy landscape $\Phi(\gamma)$ ¹⁸,

$$\frac{\partial \rho(\gamma, t)}{\partial t} = \frac{\partial}{\partial \gamma} D \left[\frac{\partial \rho(\gamma, t)}{\partial \gamma} + \frac{\rho(\gamma, t)}{k_B T} \frac{\partial \Phi(\gamma)}{\partial \gamma} \right]. \quad (1)$$

In the simplest case, $\Phi(\gamma)$ would be just a bistable potential. In the previous equation, $\rho(\gamma, t)$ is the probability distribution function which depends on the order parameter or reaction coordinate γ , D is the diffusion coefficient, T is the temperature of the bath and k_B the Boltzmann constant. When the height of the barrier separating the two minima of the potential is large enough compared to thermal energy the systems achieves a state of quasi-equilibrium in each well. The evolution of the system then proceeds by jumps from one well to the other undergoing an activated process. The dynamics corresponding to this situation can be obtained by eliminating the fast degrees of freedom in such a way that it can

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be characterized simply by the populations at each well. The Fokker-Planck equation then reduces to the following kinetic equations governing the population dynamics at both wells^{1,19}

$$\frac{dn_1}{dt} = -\frac{dn_2}{dt} = -j(t) - j^r(t). \quad (2)$$

where the current $j(t)$ is defined as:

$$j(t) = j_{\rightarrow} - j_{\leftarrow} = (k_{\rightarrow}n_1 - k_{\leftarrow}n_2) \quad (3)$$

Here $n_1(t)$ and $n_2(t)$ are the populations at each well and $k_{\rightarrow, \leftarrow}$ are the forward and backward reaction rates given by

$$k_{\rightarrow, \leftarrow} = \frac{D\sqrt{\Phi''(\gamma_{1,2})|\Phi''(\gamma_0)|}}{2\pi k_B T} \exp\left[\frac{\Phi(\gamma_{1,2}) - \Phi(\gamma_0)}{k_B T}\right], \quad (4)$$

where γ_1 and γ_2 denote the position of the two minima and γ_0 the position of the top at the barrier. The noise term $j^r(t)$ results from the coarsening of the corresponding random term $J^r(\gamma, t)$ in the Langevin equation related to the Fokker-Planck equation (1),¹. Whereas the latter satisfies the fluctuation-dissipation theorem inherent to the diffusion process along the reaction coordinate formulated as²⁰

$$\langle J^r(\gamma, t) J^r(\gamma', t') \rangle = 2D \langle \rho(\gamma, t) \rangle \delta(\gamma - \gamma') \delta(t - t') \quad (5)$$

the former does not obey a similar expression. Its correlation is given by¹⁸:

$$\langle j^r(t) j^r(t') \rangle = (k_{\rightarrow} \langle n_1 \rangle + k_{\leftarrow} \langle n_2 \rangle) \delta(t - t') \neq 2k_{\rightarrow} n_1^{eq} \delta(t - t') \quad (6)$$

where the last term is the value of the correlation at equilibrium, and n_1^{eq} is the equilibrium population in the first well.

This result clearly indicates that the violation of the fluctuation-dissipation theorem is precisely due to the coarsening of the description. When one eliminates the fast variables reducing the dynamics to that of an activated process the system cannot progressively pass through local equilibrium states from one well to the other but proceeds by jumps and consequently remains outside equilibrium. This explains why the fluctuation-dissipation theorem, a result strictly valid when the fluctuations take place around an equilibrium state²¹, is not fulfilled.

From the model proposed we can analyze the non-equilibrium response of the system. Let us assume an external perturbation $-\varepsilon(t)O(t)$ that is plugged in at the waiting time t_w , defined as the time elapsed after quenching ($O(t)$ is the observable considered and

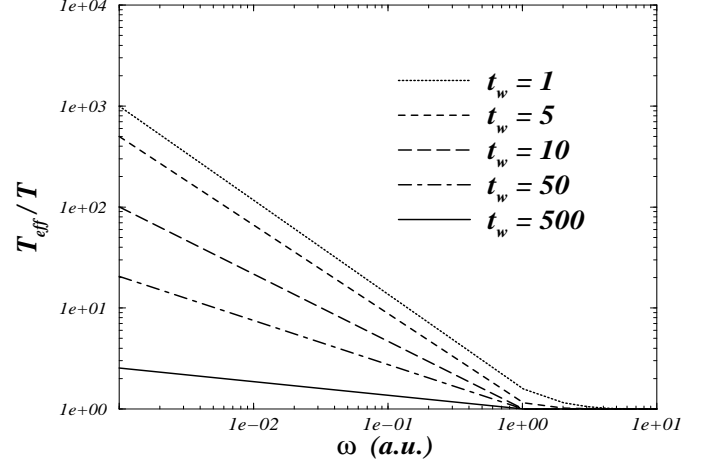


FIG. 1: The effective temperature plotted as a function of the frequency (in arbitrary units) for different values of the waiting time.

$\varepsilon(t) = \varepsilon_0 \theta(t - t_w)$ is a generalized force that is coupled to the observable). The response is then given by

$$R(t, t_w) = \left. \frac{\partial \langle \delta O(t) \rangle}{\partial \varepsilon(t_w)} \right|_{\varepsilon_0 \rightarrow 0}, \quad (7)$$

where the average of the observable is defined as

$$\langle \delta O(t) \rangle = \int O(\gamma) \delta \rho(\gamma, t) d\gamma = (O_1 - O_2) \delta n_1, \quad (8)$$

whereas the values O_1 and O_2 are the values of the observable at the minima 1 and 2, respectively. On the other hand, the correlation is

$$C_O(t, t_w) = e^{-(t-t_w)/\tau} (O_1 - O_2) [j_{\rightarrow}(O_1 - O_0) - j_{\leftarrow}(O_2 - O_0)]. \quad (9)$$

Both quantities satisfy the relation

$$R_O(t, t_w) = \frac{1}{k_B T_{eff}^O} \frac{\partial}{\partial t_w} C_O(t, t_w), \quad (10)$$

in which the quantity T_{eff}^O plays the role of an “effective” temperature given by

$$T_{eff}^O = \frac{T}{A e^{-t_w/\tau} + (1 - e^{-t_w/\tau})}. \quad (11)$$

Here $\tau^{-1} = (k_{\rightarrow} + k_{\leftarrow})$ is the relaxation time of the process and the parameter A has the form

$$A = \frac{k_{\rightarrow} \langle n_1(0) \rangle (O_1 - O_0) - k_{\leftarrow} \langle n_2(0) \rangle (O_2 - O_0)}{k_{\rightarrow} n_1^{eq} (O_1 - O_2)} \quad (12)$$

Since the equilibrium state is achieved for $t_w \rightarrow \infty$, one can easily verify that the effective temperature coincides with that of the bath and the relation (10) becomes the fluctuation-dissipation theorem.

An important consequence of our previous analysis is that the effective temperature is not a robust quantity since it depends on the observable considered as well as on the initial populations at the wells. We then conclude that in the case of an activated process that quantity is a parameter measuring the distance of the system to the equilibrium state but it is not universal and consequently can hardly be considered as a thermodynamic temperature¹⁰.

Our expression Eq.(11) can be used to reproduce the dependence of the violation factor on the frequency observed in experiments^{11,12}. If we assume that the whole relaxation process takes place through consecutive activated processes for which Eq.(2) applies, we can infer a global behavior of that quantity by identifying the inverse of the relaxation time with a frequency. By considering the case $A = 0$, in which the violation factor does not depend on the observable, we then conclude that the effective temperature at low frequencies behaves as: $T_{eff} \sim \omega^{-1}$. In the experiments one finds that the effective temperature decreases when increasing the frequency following a power law whose exponent is in between -1.1 and -1.2 ,¹². As in the experiments, it is found that the effective temperature tends to the bath temperature for very large waiting times (see Fig. 1).

III. INTERMITTENT DYNAMICS

We will analyze in this section the intermittent behavior of the fluctuations taking place when the system is quenched below the glass transition. This type of behavior has been reported in experiments measuring the fluctuation spectrum and the response of a Laponite solution and of a polymer glass^{11,12}. To this purpose, we will particularize the model discussed in Sec. 2 to the case of the quartic potential plotted in Fig. 2. The dynamics of this model is governed by the Fokker-Planck equation (1) or through the equivalent Langevin equation

$$\frac{d\gamma}{dt} = -\gamma(\gamma - \gamma_1)(\gamma - \gamma_2) + J^r. \quad (13)$$

where J^r has been considered a Gaussian white noise process having, as a first approximation, a constant amplitude: $D\langle\rho(\gamma, t)\rangle \approx \alpha$. We will see that this approximation is enough to explain the experimental results.

By numerical simulation of the Langevin equation (13), we have computed the value of the following observable

$$O(\gamma) = \begin{cases} -a_1(\gamma - \gamma_1) & \text{if } \gamma < 1 \\ a_2(\gamma - \gamma_2) & \text{if } \gamma > 1 \end{cases}, \quad (14)$$

which has a piece-wise dependence on γ , having a zero value at γ_1 and γ_2 . In our case we have taken $\gamma_1 = -2$

and $\gamma_2 = 3$ and the parameters a_1 and a_2 have been chosen arbitrarily as $a_1 = 6$ and $a_2 = 9$, ensuring the continuity of the observable. The location of the maximum of the observable has been placed at $\gamma = 1$, to show in a clearer way the occurrence of a transition between the two wells. When the state is at the top of the barrier the probability to go back to the minimum γ_1 is 50%.

The value of α should be consistent with the non-stationary nature of the process and has to be intrinsically related to the waiting time. Therefore, we have used different values of this parameter to mimic different values of the waiting time.

In Fig. III we have represented the value of the observable $O(\gamma)$ corresponding to a single trajectory obtained from the simulations of Eq.(13), with $\alpha = 1.5$. We observe the presence of an intermittent event, which is the signature of a jump from one well to another. Therefore, the presence of intermitencies is a consequence of the activated nature of the relaxation process.

Averaging over many trajectories we have obtained the probability distribution function for the observable, which has been plotted in Fig. 4, for different values of α . The non-Gaussian form of the curves is, in our model, a consequence of the non-parabolic form of the potential. Intermittencies add more weight to the tails of the distribution, thus stressing its non-Gaussian nature.

IV. CONCLUSIONS

In this paper we have used the activation over a barrier to model the evolution of a system when is quenched below the glass transition. Using as a model a simple quartic potential, we have proved the existence of an intermittent dynamics in the aging process caused by the presence of large fluctuations. Similar behavior has been observed in recent experiments. We have shown that the

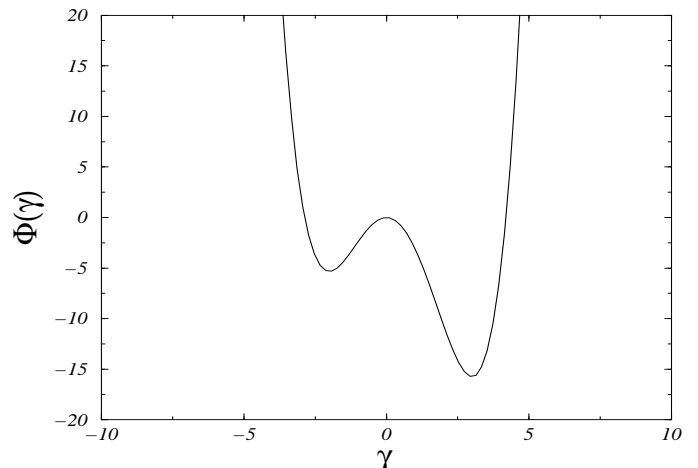


FIG. 2: Quartic potential used in our model.

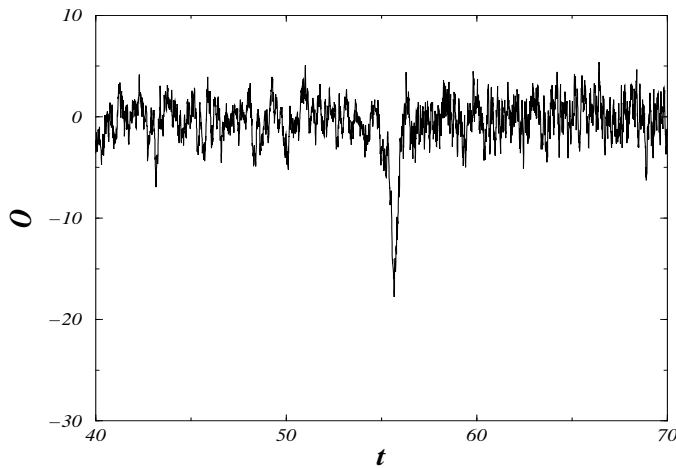


FIG. 3: The evolution of a single realization of the observable $O(\gamma)$, corresponding to $\alpha = 1.5$. The single step-time has been taken as 0.01. We have considered 10000 time intervals.

probability distribution function corresponding to those events deviates from its Gaussian form observed for systems close to equilibrium and exhibits exponential tails as those encountered experimentally.

The model proposed and its possible generalizations could, with the help of the stochastic processes theory^{22,23}, constitute a useful tool to characterize the dynamics of systems far from equilibrium for which a common phenomenology in the behavior of the fluctua-

tions is being found.

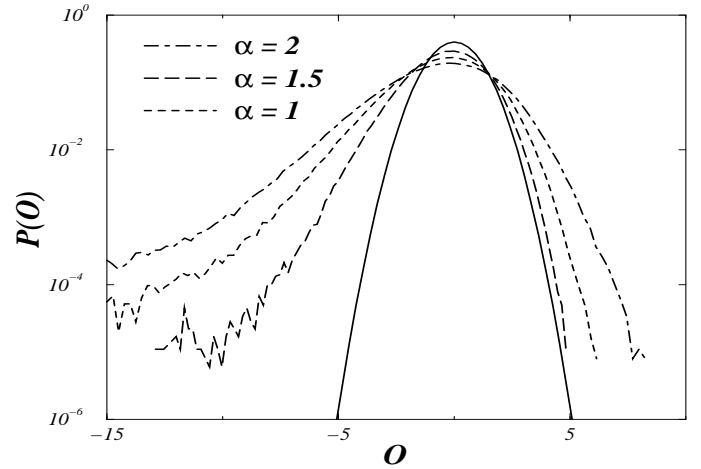


FIG. 4: Probability distribution function for different values of α as a function of the values of the observable, obtained by performing simulation of Eq.(13). The solid line is a Gaussian curve plotted just for comparison.

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